

Cavity Quantum Electrodynamics with a Single Quantum Dot Coupled to a Photonic Molecule

Arka Majumdar,^{*} Armand Rundquist, Michal Bajcsy, and Jelena Vučković
*E.L.Ginzton Laboratory,
Stanford University, Stanford, CA, 94305*

We demonstrate the effects of cavity quantum electrodynamics for a quantum dot coupled to a photonic molecule, consisting of a pair of coupled photonic crystal cavities. We show anti-crossing between the quantum dot and the two super-modes of the photonic molecule, signifying achievement of the strong coupling regime. From the anti-crossing data, we estimate the contributions of both mode-coupling and intrinsic detuning to the total detuning between the super-modes. Finally, we also show signatures of off-resonant cavity-cavity interaction in the photonic molecule.

A single quantum dot (QD) coupled to a photonic crystal (PC) cavity is an important building block for integrated nanophotonic quantum information processing devices [1]. This solid-state cavity quantum electrodynamic (cQED) system is of considerable interest to the quantum optics community for the generation of non-classical states of light [2, 3], for its application to all-optical [4, 5] and electro-optical switching [6], and due to unusual effects like the off-resonant dot-cavity interaction due to electron-phonon coupling [7]. However, all of the cQED effects demonstrated so far in this system involve a single cavity. Although numerous theoretical proposals employing multiple cavities coupled to quantum dots exist in the literature [8–10], experimental development in this direction is rather limited. Recently it has been reported that strongly sub-Poissonian light can be generated from a pair of coupled cavities containing a single QD [11, 12]. This double cavity, also called a photonic molecule, coupled to a single QD forms the first step towards building an integrated cavity network with coupled QDs. Photonic molecules made of PC cavities were studied previously [13, 14] to observe mode-splitting due to coupling between the cavities. In those studies, a high density of QDs was used merely as an internal light source to generate photoluminescence (PL) under above-band excitation and no quantum properties of the system were studied. In another experiment, a photonic molecule consisting of two micropost cavities was used along with a single QD to generate entangled photons via exciton-biexciton decay, but the QD-cavity system was in the weak coupling regime and the Purcell enhancement was the only cQED effect observed [15].

In this paper, we demonstrate strong coupling of a photonic molecule with a single QD. We show clear anti-crossing between the QD and two super-modes formed in the photonic molecule. In general, the exact coupling strength between two cavities in a photonic molecule is difficult to calculate, as the observed separation between the two modes has contributions both from the cavity

coupling strength as well as from the mismatch between the two cavities due to fabrication imperfections. However, by monitoring the interaction between a single QD and the photonic molecule we can exactly calculate the coupling strength between the cavities and separate the contribution of the bare detuning due to cavity mismatch. In fact, without any coupling between two cavities, one cannot have strong coupling of the QD with both of the observed modes. Hence, the observed anti-crossing of the QD with both modes clearly indicates coupling between the cavities. Apart from the strong coupling, we also demonstrate off-resonant phonon-mediated interaction between the two cavity modes, a recently found effect in solid-state cavity systems.

Let us consider a photonic molecule consisting of two cavities with annihilation operators for their bare (uncoupled) modes denoted by a and b , respectively. We assume that a QD is placed in and resonantly coupled to the cavity described by operator a . The Hamiltonian describing such a system is:

$$\mathcal{H} = \Delta_o b^\dagger b + J(a^\dagger b + ab^\dagger) + g(a^\dagger \sigma + a \sigma^\dagger) \quad (1)$$

where Δ_o is the detuning between the two bare cavity modes; J and g are, respectively, the inter-cavity and dot-cavity coupling strength; σ is the QD lowering operator; and the resonance frequency ω_0 of the cavity with annihilation operator a is assumed to be zero. We now transform this Hamiltonian by mapping the cavity modes a and b to the bosonic modes α and β introduced as $a = \cos(\theta)\alpha + \sin(\theta)\beta$ and $b = \sin(\theta)\alpha - \cos(\theta)\beta$. We note that this mapping maintains the appropriate commutation relations between operators a and b . Under these transformations we can decouple the two cavity modes (α and β) for the following choice of θ :

$$\tan(2\theta) = -\frac{2J}{\Delta_o} \quad (2)$$

Under this condition the transformed Hamiltonian becomes:

$$\begin{aligned} \mathcal{H} = & \alpha^\dagger \alpha (\Delta_o \sin^2(\theta) + J \sin(2\theta)) + g \cos(\theta) (\alpha^\dagger \sigma + \alpha \sigma^\dagger) \\ & + \beta^\dagger \beta (\Delta_o \cos^2(\theta) - J \sin(2\theta)) + g \sin(\theta) (\beta^\dagger \sigma + \beta \sigma^\dagger) \end{aligned}$$

^{*}Electronic address: arkam@stanford.edu

Therefore, a QD coupled to a photonic molecule has exactly the same eigen-structure as two detuned cavities with the QD coupled to both of them (from the equivalence of the two expressions above for the Hamiltonian \mathcal{H}). The super-modes of the transformed Hamiltonian α and β will be separated by $\Delta = \sqrt{\Delta_o^2 + 4J^2}$ (obtained by subtracting the terms multiplying $\alpha^\dagger\alpha$ and $\beta^\dagger\beta$, under the conditions of Eq.2) and the interaction strength between the QD and the super-modes will be $g_1 = g \cos(\theta)$ and $g_2 = g \sin(\theta)$. If the two cavities are not coupled ($J = 0$ and $\theta = 0$), we can still observe two different cavity modes in the experiment due to Δ_o , the intrinsic detuning between two bare cavities. However, if we tune the QD across the two cavities in this case, we will observe QD-cavity interaction only with one cavity mode (in this case α , as the term coupling β to the QD in the transformed Hamiltonian will vanish, as a result of $\sin(\theta) = 0$). In other words, in this case the QD is spatially located in only one cavity and cannot interact with the other, spatially distant and decoupled cavity. Fig. 1 shows the numerically calculated cavity transmission spectra (proportional to $\langle a^\dagger a \rangle + \langle b^\dagger b \rangle$) when the QD is tuned across the two cavity resonances. When the two cavities are coupled ($J \neq 0$), we observe anti-crossing between each cavity mode and the QD (Fig. 1a). However, only one anti-crossing is observed when the cavities are not coupled (Fig. 1b).

The actual experiments are performed with self-assembled InAs QDs embedded in GaAs, and the whole system is kept at cryogenic temperatures ($\sim 10 - 25$ K) in a helium-flow cryostat. The cavities used are linear three hole defect GaAs PC cavities coupled via spatial proximity. The photonic crystal is fabricated from a 160 nm thick GaAs membrane, grown by molecular beam epitaxy on top of a GaAs (100) wafer. A low density layer of InAs QDs is grown in the center of the membrane (80 nm beneath the surface). The GaAs membrane sits on a 918 nm sacrificial layer of $\text{Al}_{0.8}\text{Ga}_{0.2}\text{As}$. Under the sacrificial layer, a 10-period distributed Bragg reflector, consisting of a quarter-wave AlAs/GaAs stack, is used to increase collection into the objective lens. The photonic crystal was fabricated using electron beam lithography, dry plasma etching, and wet etching of the sacrificial layer in diluted hydrofluoric acid, as described previously [7, 16].

We fabricated two different types of coupled cavities: in one case, the two cavities are offset at a 30° angle (inset of Fig. 2a) and in the other the two cavities are laterally coupled (inset of Fig. 2b). In the first case the coupling between the cavities is stronger as the overlap between the electromagnetic fields confined in the cavities is larger along the 30° angle. Figs. 2a,b show the typical PL spectra of these two different types of coupled cavities for different spacing between the cavities. A clear decrease in the frequency separation between the cavities is observed with increasing spatial separation. Note that the consistency of this trend between different fabrication runs already indicates that this frequency separation

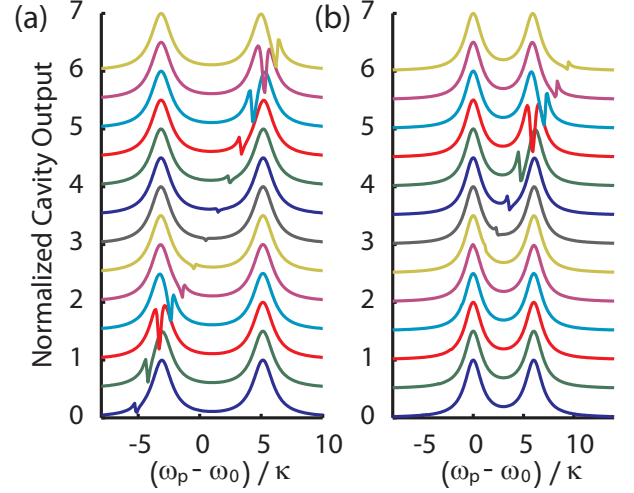


FIG. 1: (color online) Numerically calculated cavity transmission spectra when the QD resonance is tuned across the two cavity resonances. (a) Anticrossing is observed between the quantum dot and both cavity modes when the two cavities are coupled (coupling rate between the two cavities is $J/2\pi = 80$ GHz). (b) When the two cavities are not coupled ($J = 0$), we observe anti-crossing in only one cavity. Parameters used for the simulation: cavity decay rate $\kappa/2\pi = 20$ GHz (for both cavities); QD dipole decay rate $\gamma/2\pi = 1$ GHz; dot-cavity coupling rate of $g/2\pi = 10$ GHz; intrinsic detuning between the bare cavity modes $\Delta_o/2\pi = 40$ GHz for (a) and 120 GHz for (b). The plots are vertically offset for clarity. The horizontal axis corresponds to the detuning of the probe laser frequency ω_p from the cavity a resonance ω_0 in units of cavity field decay rate.

cannot be purely due to the fabrication-related intrinsic detuning between the two cavities. Nevertheless, it is very difficult to quantify how much of the separation is due to coupling (J), and how much is due to intrinsic detuning (Δ_o) of the cavity resonances. However, we will show that by observing the anti-crossing between the QD and the two modes we can conclusively determine both J and Δ_o .

First, we investigate the strong coupling between a single QD and the photonic molecule. For this particular experiment, we used a photonic molecule consisting of cavities separated by 4 holes along the 30° angle. In practice it is not trivial to tune the QD over such a long wavelength range as required by the observed separation of the two cavity peaks. Hence we use two different tuning techniques: we tune the cavity modes by depositing nitrogen on the cavity [17], and then tune the QD resonance across the cavity resonance by changing the temperature of the system. We observe clear anti-crossings for both the modes as shown in Figs. 3a,b. Fig. 3a is obtained by temperature-tuning the QD across the longer-wavelength cavity mode before nitrogen deposition. We then perform the nitrogen deposition to red-shift the cavity resonances, and repeat the temperature tuning. Fig. 3b shows the anti-crossing between the QD and the shorter-wavelength

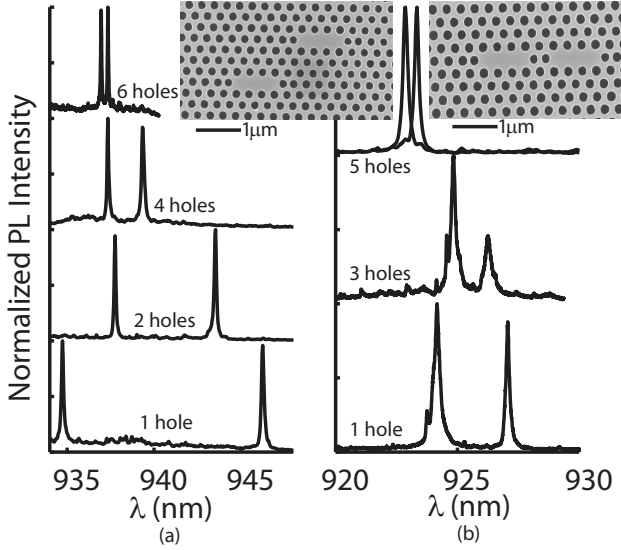


FIG. 2: Photoluminescence spectra of the coupled cavities for different hole spacings between two cavities: (a) the cavities are separated at an angle of 30° (see the inset for a scanning electron micrograph (SEM)); (b) the cavities are laterally separated (see the inset for SEM). A decrease in the wavelength separation between two cavity modes is observed with increasing spatial separation between the cavities (i.e., with increasing number of holes inserted in between the two cavities). A much larger separation is observed in (a) when the cavities are coupled at an angle compared to the lateral coupling (b).

cavity mode. The nitrogen and the temperature tuning do not cause a significant change in the coupling and the detuning between the cavities, as confirmed in the experiments described below.

We perform curve-fitting for the PL spectra when the QD is resonant to the cavity super-modes and estimate the system parameters (Figs. 4a,b). The super-mode at shorter (longer) wavelength is denoted as sm1 (sm2). As the detuning between the super-modes is much larger than the vacuum Rabi splitting caused by the QD, we can assume that when the QD is resonant to sm1(2), its interaction with sm2(1) is negligible. Therefore, we can fit the PL spectra of sm1 (sm2) modes exhibiting Rabi splitting individually. For sm1, we extract from the fit the field decay rate $\kappa_1/2\pi = 16.7$ GHz and the QD-field interaction strength $g_1/2\pi = 23.7$ GHz (Fig. 4a); for sm2, $\kappa_2/2\pi = 22.4$ GHz and $g_2/2\pi = 14.2$ GHz (Fig. 4b). We note that we can achieve very high quality factors ($\sim 7,000 - 10,000$) of the coupled cavity modes as seen from the extracted κ values. We also estimate the total detuning between two observed modes as $\Delta/2\pi = 0.7$ and 0.72 GHz before and after nitrogen tuning. This minimal difference in Δ resulting from the nitrogen tuning does not impact our further analysis, and we take Δ to be the average of these two values. The change in the cavity field decay rates arising from the nitrogen deposition is also minimal. From these data, we use the relations $\theta = \arctan(g_2/g_1)$, $\tan(2\theta) = -2J/\Delta_o$ and $\Delta = \sqrt{4J^2 + \Delta_o^2}$

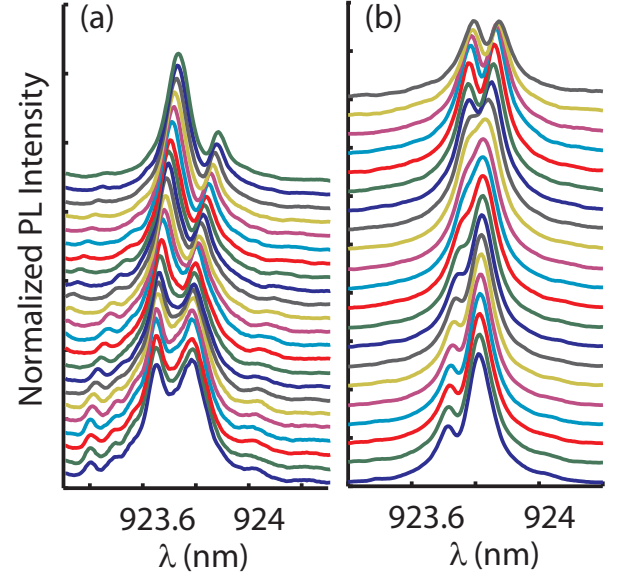


FIG. 3: (color online) Normalized PL intensity plotted when we tune the QD across the cavity resonance by temperature: (a) before nitrogen deposition (i.e., the QD is temperature tuned across the longer wavelength resonance), and (b) after nitrogen deposition (which red-shifts the cavity resonances and allows us to temperature tune the QD across the shorter wavelength resonance). Clear anti-crossings between the QD and the cavity are observed for both super-modes. In both cases, the temperature is increased from top to bottom (the plots are vertically offset for clarity).

to obtain: $J/2\pi \approx 110$ GHz and $\Delta_o/2\pi \approx 118$ GHz.

We now numerically simulate the performance of such a QD-photonic molecule for generation of sub-Poissonian light using the quantum optical master equation approach [18]. Two bare cavity modes are separated by $\Delta_o/2\pi = 118$ GHz; a QD is resonant and strongly coupled to one of the modes (a) with interaction strength $g/2\pi = 27.6$ GHz ($g = \sqrt{g_1^2 + g_2^2}$, where g_1 and g_2 are the two values of QD-cavity interaction strengths obtained by fitting the PL spectra); mode *b* is the empty cavity. The mode *b* is driven and the second order autocorrelation $g^2(0) = \frac{\langle b^\dagger b^\dagger b b \rangle}{\langle b^\dagger b \rangle^2}$ of the transmitted light through cavity *b* is calculated [12]. We also assume the two cavities to have the same cavity decay rate, which is an average of the cavity decay rates measured from the two super-modes. Note however that, having slightly different decay rates does not significantly affect the performance of the system. The numerically simulated cavity *b* transmission and $g^2(0)$ of the transmitted light is shown in Figs. 4c,d. We note that with our system parameters we can achieve strongly sub-Poissonian light with $g^2(0) \sim 0.03$. Unfortunately, in practice it is very difficult to drive only one cavity mode without affecting the other mode due to the spatial proximity of two cavities. This individual addressability is critical for good performance of the system [12] and to retain such a capability in a photonic

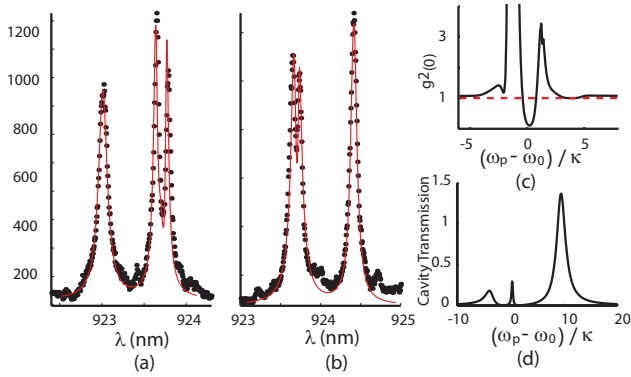


FIG. 4: (color online) QD-photonic molecule spectrum, (a) when the QD is resonant with super-mode sm1 and (b) when the QD is resonant with super-mode sm2. From the fit we extract the system parameters (see text). Numerically simulated (c) second order autocorrelation $g^2(0)$ and (d) transmission from cavity b , as a function of laser frequency, with the experimental system parameters that were extracted from the fits.

molecule the cavities should be coupled via a waveguide [19].

Finally, as a further demonstration of cQED effects in this system, we report off-resonant interaction between the coupled cavities and the QD, similar to the observations in a single linear three hole defect cavity [20] and a nano-beam cavity [21]. This experiment was performed on a different QD-photonic molecule system than the one where we observed strong coupling. Fig. 5 shows the spectra indicating off-resonant coupling between the cavities and the QD. Under resonant excitation of the supermode at longer wavelength (sm2), we see pronounced emission from both sm1 and a nearby QD. Similarly, under resonant excitation of sm1, we see emission from sm2, although the emission is much weaker. We exclude the presence of any nonlinear optical processes by performing a laser-power dependent study of the cavity emission, which shows a linear dependence of the cavity emission on the laser power (not shown here).

In summary, we demonstrated strong coupling of a single QD to a photonic molecule in a photonic crys-

tal platform. Clear anti-crossings between the QD and both super-modes of the photonic molecule were observed, showing conclusive evidence of inter-cavity coupling. From the anti-crossing data we were able to separate the contributions of the inter-cavity coupling and intrinsic detuning to the cavity mode splitting. We have also reported observation of off-resonant cavity-cavity and cavity-QD interaction in this type of system. Such a system could be employed for non-classical light generation (as theoretically studied in this article), and represents a building block for an integrated nanophotonic network in a solid-state cQED platform.

The authors acknowledge financial support provided by the Office of Naval Research (PECASE Award; No: N00014-08-1-0561), DARPA (Award No: N66001-12-1-

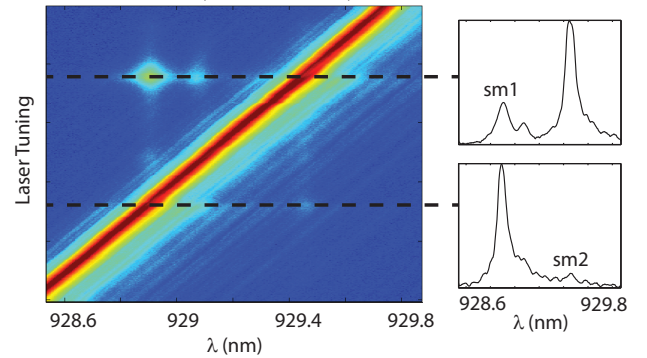


FIG. 5: (color online) Off-resonant interaction between two coupled cavities and a QD. We scan the laser across both coupled modes, and observe emission from the off-resonant super-mode, under excitation of the other super-mode. A close-up spectrum for each resonance shows the relative position of the laser and the cavity modes.

4011), NSF (DMR-0757112) and Army Research Office (W911NF-08-1-0399). A.R. is also supported by a Stanford Graduate Fellowship. We acknowledge Dr. Hyochul Kim and Dr. Pierre Petroff for providing the quantum dot sample. This work was performed in part at the Stanford Nanofabrication Facility of NNIN, supported by the National Science Foundation.

- [1] A. Faraon, A. Majumdar, D. Englund, E. Kim, M. Bajcsy, and J. Vučković, *New J. Physics* **13**, 055025 (2011).
- [2] A. Faraon, I. Fushman, D. Englund, N. Stoltz, P. Petroff, and J. Vučković, *Nature Physics* **4**, 859 (2008).
- [3] A. Majumdar, M. Bajcsy, and J. Vučković, arXiv:1106.1926 (2011).
- [4] D. Englund, A. Majumdar, M. Bajcsy, A. Faraon, P. Petroff, and J. Vučković, arXiv:1107.2956 (2011).
- [5] D. Sridharan, R. Bose, H. Kim, G. S. Solomon, and E. Waks, arXiv:1107.3751v1 (2011).
- [6] A. Faraon, A. Majumdar, H. Kim, P. Petroff, and J. Vučković, *Phys. Rev. Lett.* **104**, 047402 (2010), URL

<http://link.aps.org/doi/10.1103/PhysRevLett.104.047402>.

- [7] D. Englund, A. Majumdar, A. Faraon, M. Toishi, N. Stoltz, P. Petroff, and J. Vučković, *Phys. Rev. Lett.* **104**, 073904 (2010).
- [8] M. J. Hartmann, F. G. S. L. Brandao, and M. B. Plenio, *Nature Physics* **2**, 849 (2006).
- [9] A. D. Greentree, C. Tahan, J. H. Cole, and L. C. L. Hollenberg, *Nature Physics* **2**, 856 (2006).
- [10] I. Carusotto, D. Gerace, H. E. Tureci, S. De Liberato, C. Ciuti, and A. Imamoglu, *Phys. Rev. Lett.* **103**, 033601 (2009).

- [11] T. C. H. Liew and V. Savona, Phys. Rev. Lett. **104**, 183601 (2010), URL <http://link.aps.org/doi/10.1103/PhysRevLett.104.183601>.
- [12] M. Bamba, A. Imamoğlu, I. Carusotto, and C. Ciuti, Phys. Rev. A **83**, 021802 (2011), URL <http://link.aps.org/doi/10.1103/PhysRevA.83.021802>.
- [13] K. A. Atlasov, K. F. Karlsson, A. Rudra, B. Dwir, and E. Kapon, Optics Express **16**, 16255 (2008).
- [14] K. A. Atlasov, A. Rudra, B. Dwir, and E. Kapon, Optics Express **19**, 2619 (2011).
- [15] A. Dousse, J. Suffczynski, A. Beveratos, O. Krebs, A. Lematre, I. Sagnes, J. Bloch, P. Voisin, and P. Senellart, Nature **466**, 217220 (2010).
- [16] D. Englund, A. Faraon, I. Fushman, N. Stoltz, P. Petroff, and J. Vučković, Nature **450**, 857 (2007).
- [17] S. Mosor, J. Hendrickson, B. C. Richards, J. Sweet, G. Khitrova, H. M. Gibbs, T. Yoshie, A. Scherer, O. B. Shchekin, and D. G. Deppe, Applied Physics Letters **87**, 141105 (pages 3) (2005), URL <http://link.aip.org/link/?APL/87/141105/1>.
- [18] A. Majumdar, E. Kim, Y. Gong, M. Bajcsy, and J. Vučković, Phys. Rev. B **84** (2011).
- [19] Y. Sato, Y. Tanaka, J. Upham, Y. Takahashi, T. Asano, and S. Noda, Nature Photonics **6**, 5661 (2012).
- [20] A. Majumdar, A. Faraon, E. D. Kim, D. Englund, H. Kim, P. Petroff, and J. Vučković, Phys. Rev. B **82**, 045306 (2010).
- [21] A. Rundquist, A. Majumdar, and J. Vučković, Applied Physics Letters **99**, 251907 (pages 3) (2011), URL <http://link.aip.org/link/?APL/99/251907/1>.